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Particle Size Fractionation of Airborne Gamma-Emitting Radionuclides by Graded Filters

Abstract. *Three filters in series were employed to separate into groups, according to size, the nuclear debris suspended in air at ground level. The concentrations of airborne Ce^{144}-Pr^{144}, Sb^{125}, Cs^{137}, Zr^{95}-Nb^{95}, Mn^{54}, and Ru^{106}-Rh^{106} in each size group were determined by gamma spectral analysis. Particles having diameters of less than 1.75 microns contained a minimum of 88 percent of the total activity.*

The particle size distribution of airborne radioactivity is important because it influences the rates of sedimentation, of removal from the atmosphere by precipitation, and of ground deposition of fallout. By determining the concentration of individual radionuclides in fractions of specific particle size the physical factors operative in isotopic fractionation and in the transport of nuclear debris in the atmosphere can be elucidated. Furthermore, knowledge of the particle size distribution of airborne radionuclides is pertinent to the estimation of the inhalation hazard because retention of particulate material in the lungs is not a sole function of the quantity inhaled, but also varies with particle size distribution, solubility, and aerodynamic properties.

In our study we used three filters of increasing retentivities, in series, to separate airborne particles into size groups. The radionuclide composition of the material retained on each filter was determined by gamma spectral analysis and the distribution calculated for each nuclide (Ce^{144}-Pr^{144}, Sb^{125}, Cs^{137}, Zr^{95}-Nb^{95}, Mn^{54}, and Ru^{106}-Rh^{106}) on the three filters.

Table 1. Average particle size parameters. The geometric S.D. (standard deviation) is the ratio of the 84-percent diameter to the 50-percent diameter.

		Filter A		Filter B		Filter C	
Sample duration (days)	No. of samples averaged	Geometric mean diameter (μ)	Geometric S.D.	Geometric mean diameter (μ)	Geometric S.D.	Geometric mean diameter (μ)	Geometric S.D.
1	4	0.65	2.12	0.36	1.80	0.28	1.57
3	7	.87	1.83	.42	1.84	.25	1.81
5	3	.89	2.11	.48	1.77	.28	1.70

Table 2. Fraction of total radionuclide activity collected on each filter.

No. of samples averaged*	Nuclide	Filter A		Filter B		Filter C	
		Range	Average	Range	Average	Range	Average
19	Ce^{144}-Pr^{144}	0.07–0.19	0.11	0.59–0.78	0.73	0.12–0.26	0.16
18	Sb^{125}	.07– .22	.11	.58– .78	.72	.10– .25	.17
8	Cs^{137}	.08– .19	.12	.65– .80	.72	.12– .22	.16
19	Zr^{95}-Nb^{95}	.06– .20	.12	.61– .77	.72	.10– .24	.16
19	Mn^{54}	.06– .25	.11	.57– .80	.74	.08– .24	.15
19	Ru^{166}-Rh^{106}	.07– .24	.13	.57– .78	.71	.10– .23	.16

* Because of low activity in some instances, less than 19 samples were averaged.

Jet impactors and high-speed centrifugal precipitators were used in previous studies of this kind. While such devices have the advantage of separating particles on the basis of their aerodynamic properties, they tend to break up lose aggregates of particles and their low sampling rates make determination of low-levels of radioactivity impractical. Two-stage samplers with a small cyclone separator to remove nonrespirable particles somewhat overcome the disadvantage of low sampling rates but risk the loss of potentially useful information on particle size. As an alternative to such techniques, it was suggested that differences in filter retentivities might provide a means for studying airborne particle size distribution (*1*). The individual radionuclides obtained on filters can be quantified by radiochemical and gamma analysis. The advantage of this technique is that it permits the collection of air samples of large volume, the determination of particle size, and the non-destructive radionuclide analysis of the collected fission products.

We used a sampling head consisting of three graded 20- by 25-cm filters (referred to as filters A, B, and C) and probes up- and down-stream of each graded filter to sample a portion of the air stream. A rotary blower provided a sampling rate of 140 liters per minute through the system. Filter media CM-245 (designated filter A) (*2*), CM-285 (designated filter B), and MSA-1106BH (designated filter C) were selected from many filters tested because of their different retentivities of airborne Zr^{95}-Nb^{95}, similar gamma-counting efficiencies, and similar glass-fiber composition. The selected filters differed from those employed in previous studies by other investigators (*3*).

To obtain the relative number of particles in a specific size range captured by each graded filter, samples collected by means of the up- and down-stream probes on type AA membrane filters (*2*) were optically sized at 100× and 430×, and at oil immersion magnification (×1000). A minimum of 150 particles, or the particles distributed over an area of 3200 μ^2, were sized; although in most cases twice the minimum number of particles were sized for each probe sample. Limitations inherent in optical microscopic examination, particularly the inability to detect particles smaller than approximately 0.3 μ, tend to shift size distributions obtained.

Differences in the probe counts yielded the relative number of particles captured by the graded filter between them. If a logarithmic distribution for the size of particles present is assumed, the cumulative percentage of particles in a size range plotted against the center of the size range in microns can be used to characterize the distribution. The diameter at which half the number of particles have diameters greater than the center of the size range and half have diameters less was designated the geometric mean diameter. The geometric standard deviation was thus the ratio of the 84-percent diameter to the 50-percent diameter.

The graded filters, folded in quarters, were counted for gamma radioactivity for a minimum of 900 minutes on a 10- by 10-cm NaI crystal in a steel shield 15 cm thick (*4*). The activities of each radionuclide on the filter were determined by quantifying the gamma spectrum by a method which determines the net counts in the pertinent peaks by subtraction of background gamma radiation, by application of multilinear equations to correct for mutual interferences of the radionuclides, and by application of appropriate constants to convert counts per minute to picocuries (*5*).

To estimate the effective age of origin of the fission products captured, the ratio of Zr^{95}-Nb^{95} to gross beta activity was employed. From curves of fission yields the effective age of the material was then estimated (*6*). Counting errors alone limit this estimation to ±20 days from 400-day-old material.

Samples collected over 1, 3, and 5 days at a constant linear air velocity of 60 cm/sec were obtained between 9 March 1964 and 20 May 1964. The sampler was situated in a non-urban area approximately 2 meters above ground level. The amount of particulate material collected on the filters varied from 60 mg collected during 24 hours to 476 mg collected over 5 days of sampling. The airborne particles entering the sampler had a geometric mean diameter of 0.32 to 0.41 μ and a geometric standard deviation ranging from 1.80 to 2.06. The effective age of origin of the fission products sampled varied from 440 to 550 days.

The average geometric mean diameters and standard deviations for microscopically sized samples are presented in Table 1. In no case did the probe behind filter C show any evidence of particles. The greatest variations in particle size parameters occurred on filter A. Filter B captured particles having essentially the same geometric mean diameter as particles present in the surrounding air. From the average values in Table 1, distribution curves of particle size were plotted on log-probability graph paper. The percentage of the total number of particles in each interval was plotted in histogram form (Fig. 1) with convenient particle size intervals (0.25 μ). Variations in the percentage of particles retained in any size interval from samples collected over 1 to 5 days did not exceed 15 percent and in most instances were not greater than 10 percent. This variation is due to the interplay of differences in the particle size distribution of the incoming aerosol, the loading of the filters, and the duration of sampling. All particles collected on filters C and B had diameters of less than 1.5 μ and 1.75 μ, respectively. Filter A removed all particles greater than 1.75 μ but contained particles throughout the entire size range.

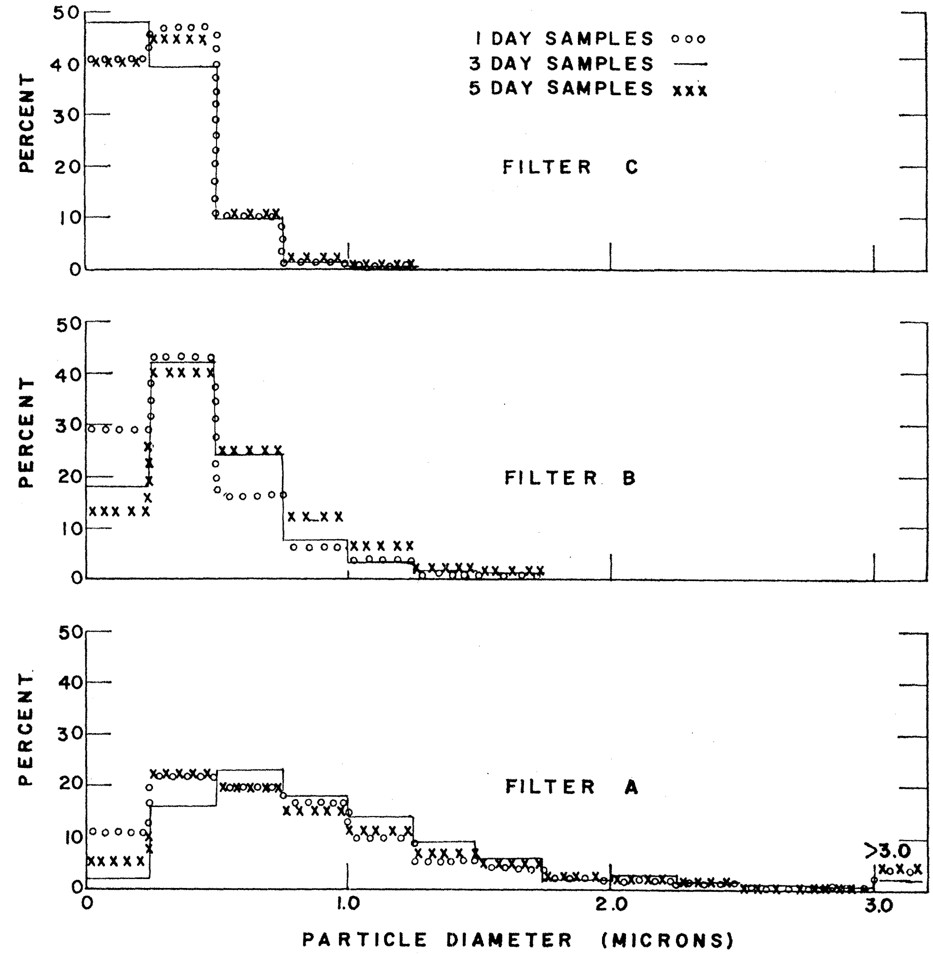

Fig. 1. Histogram of particle size distributions based on average geometric mean diameters and geometric standard deviations for samples collected over 1, 3, and 5 days.

The fraction of the total activity captured on each filter for the fission products emitting gamma radiation is shown in Table 2. The range for the fraction of individual nuclides retained on individual filters was fairly broad, but average values were in good agreement. Fractional distributions for individual radionuclides were similar on any one sample but varied from sample to sample, possibly because of meteorological variations. Based on average values from filters B and C (Table 2), a minimum of 88 percent of the airborne particles emitting gamma radiation was associated with particles having diameters less than 1.75 μ. At least 70 percent of the radioactivity was associated with particles having a geometric mean diameter essentially the same as the airborne dust in the area.

The data are not directly comparable to other studies because of differences in effective age of origin of the fission products sampled, in the particle size distribution of the incoming aerosol, and in the particle size intervals employed (*7*). If it is assumed that the particles remain in the troposphere for 30 days (*8*), the effective age of the radioactive material in this study indicates that it originated in the stratosphere. This study indicates that long-lived fission-product activity in air at ground level is for the most part associated with airborne material having the same diameter as particles prevalent in the surrounding air. Studies of stratospheric activity show such activity to be associated with particles of smaller size (*9*). Furthermore, the long-lived nuclides present that emit gamma-radioactivity show the same particle size distribution, indicating that the dynamics of transfer of activity from the stratosphere act similarly on these six radionuclides.

There were no correlations between surface area, weight, and relative number of particles and the activity collected on a filter. This could be because the particles containing radioactivity amount to a small percentage of the total number of particles present. Particle size separations were not sufficiently distinct to determine accurately deposition in the pulmonary spaces. However, if particle densities from 1.0 to 2.0 are assumed, pulmonary deposition would vary from 25 to 60 percent for the majority of radioactive particles in this study (*10*).

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Quartz: Anomalous Weakness of Synthetic Crystals

Abstract. *The strength of a synthetic quartz crystal drops rapidly at 400°C, and at 600°C is a hundredfold lower than at 300°C. Large plastic deformations can be produced without fracture. The predominant mechanism of deformation is translation gliding. The preferred explanation for this anomalous weakness is that this synthetic quartz contains water which has hydrolyzed the silicon-oxygen bonds. The silanol groups so formed are presumed to be rendered sufficiently mobile by elevating the temperature to 400°C so that they align themselves in dislocation lines and move through the crystal with the dislocation under the small applied shear stress.*

Under appropriate circumstances synthetic quartz crystals can be molded like putty. If silicates in general can be molded in similar fashion, it might be possible to form brittle silicate materials at considerably lower temperatures than those now used in glass technology.

Natural quartz is one of the strongest of common substances, rivaling tool steel in its compressive strength of 20 kilobars (*1*). Its strength is increased under confining pressure so much that at one time Bridgman seriously considered using quartz single crystals for the second-stage pistons of his high-pressure apparatus (*2*). Despite many attempts, quartz is the only material which has not been rendered ductile by high confining pressure at room temperature (*2, 3*). Quartz crystals retain high hardness and high strength to temperatures of 800° to 1000°C at low confining pressure (*4*).

At elevated temperatures and moderate confining pressures, quartz single crystals and aggregates have been plastically deformed, and the mechanisms of deformation have been determined (*5*). The strength of quartz has been measured in the ductile regime as a function of temperature and strain rate, at a confining pressure of 15 kb (*6*), and the natural quartz crystals become very weak when in the presence of water at temperatures of 800°C and above. Microscopic analysis of thin sections suggests that this weakness is caused by the diffusion of water throughout the crystal, hydrolyzing the silicon-oxygen bonds. In an anhydrous environment, natural quartz crystals have high strength at least up to 1000°C, the limit of our tests.

We have now studied in an anhydrous environment a large optically clear synthetic quartz crystal donated by the Bell Telephone Laboratories. At low temperatures, specimens cut from this quartz crystal have strengths similar to those of natural crystals. However, as the temperature is increased, the strength is drastically reduced.

The transition from high strength to great weakness in this synthetic quartz is accompanied by changes in the mechanisms of deformation which are readily apparent when thin sections of specimens are viewed in polarized light. A series of experiments at a constant confining pressure of 15 kb and strain rate of 8×10^{-6} sec^{-1} will be used to illustrate this transition with increasing temperature. At the lowest temperature of the series, 200°C, the specimen broke at an indicated longitudinal differential stress $(\sigma_1 - \sigma_3)$ of 45 kb. The thin section exhibited primarily fracture (which occurred along basal planes) along with a few basal deformation lamellae. At 300°C considerable plastic flow occurred prior to fracture, and Fig. 1*A* shows this specimen, which has been shortened 6 percent by flow at a compressive stress of 42 kb before breaking at one end. The plastic deformation occurred primarily by basal slip concentrated in narrow kink bands subparallel to the quartz *c* axis. This behavior at 200° and 300°C is typical of that observed in natural quartz crystals at those temperatures. At 400°C, however, the strength was only 1 kb and the mechanism of deformation changed from basal to predominant prismatic slip (Fig. 1*B*). In natural crystals a change from basal to prismatic slip has also been observed, but at 700°C rather